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Effective combination of permanganate composite chemicals (PPC) and biological aerated filter (BAF) to pre-treat polluted drinking water source

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ABSTRACT

A hybrid pre-treatment process was developed, which integrated permanganate composite chemicals (PPC) and biological aerated filters (BAF) together to enhance pollutants removal and guarantee drinking water safety. The hybrid pre-treatment process presented a high removal rate of dissolved organic matter (DOC), ammonium and nitrite, with average removal rates at 34.92, 86.10, and 97.70%, respectively. PPC changed the structure of organic matter making it more biodegradable, thus increasing the biologically removal efficiency of organic matter and greatly decreasing the disinfection byproducts formation potential. Introduction of PPC would increase the manganese concentration, but the following BAF could reduce the manganese concentration to a satisfactory level (below 0.05 mg/L). Scanning electronic microscopy and PCR-DGGE demonstrated that biodegradation should be the main mechanism for impurities removal. Jar tests results indicated that combination of the hybrid pre-treatment process and coagulation presented higher removal of impurities, with DOC removal at 55.14%. It could be concluded that PPC and BAF could enhance the pollutants removal. The hybrid pre-treatment strategy could benefit the subsequent process by significantly reducing the contaminant loading, and it would be of great significance to upgrade the conventional water treatment process.

Keywords: Potassium permanganate composite; Biological aerated filter; Hybrid pre-treatment process; Polluted source water; Conventional drinking water process

1. Introduction

It is difficult for conventional water treatment processes to produce high-quality drinking water from polluted source water, as the polluted source water usually contains high concentrations of organic matter, ammonia, and other refractory impurities. These pollutants could result from the untreated wastewater discharge, but have always been major problems for conventional drinking water treatment processes. Moreover, the presence of organic matter and ammonia resulted in more demand of chlorine during the

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pre-oxidation or disinfection period, and thus would increase the disinfection byproducts formation [1].

To remove organic matter and ammonia, pre-oxidation has been extensively studied to pre-treat source water for drinking water production all over the world. Chlorine [2], ozone [3], and potassium permanganate composite (PPC) [4] are widely employed during the pre-oxidation process. Generally, dosing chlorine could effectively reduce the organic matter and ammonia in raw water, but it would greatly increase the formation potential of disinfection byproducts [5]. Though ozone presents higher oxidation capacity, it would produce some hazardous substances, such as aldehydes and bromate, which could directly influence the chemical safety of the treated water [6], and the application of ozone needs higher cost and more complex management. Pre-oxidation by PPC has been demonstrated as an efficient pretreatment technology with simple operation and low cost and a better method to control the formation potential of disinfection byproducts [7]. However, low dosage of PPC could not achieve expected removal of impurities, and the disadvantage of higher dosage was the possibility of high residual manganese concentration. On the other hand, pre-oxidation with PPC has a weakness for ammonia removal.

Therefore, an alternative choice was to combine the PPC oxidation process with other technology. Biological methods would be of great interest thanks to its green characteristics [8]. More importantly, owing to the removal of ammonia and organic matter, biological pre-filtration had been demonstrated that it could effectively reduce the following chlorine dosage in conventional drinking water plant [9]. Among the biological methods, biological aerated filter (BAF) has been widely used in the wastewater treatment plant for its small footprint and high performance [10]. BAF process has been reported to be efficient to remove ammonia, organic matter, and manganese from the feed water [11], and had been widely studied with various filter media [12,13]. Moreover, BAF was also an effective method to handle ammonia and organic matter in drinking water treatment [14,15]. According to our previous study, lava rock could act well in BAF, and to our knowledge, little research on the performance of lava-based BAF has hardly been reported for drinking water production, comparing to other kind of biological processes, such as biological activated carbon. However, biological methods as terminal process would release filter fines and/or bacteria to the drinking water distribution system [16].

Based on the hypothesis that PPC and lava-based BAF could complement each other for contaminants removal, the objective of this investigation was to evaluate the feasibility to combine PPC pre-oxidation and lava-based BAF as the pre-treatment process of polluted source water before coagulation. The performance of the hybrid pre-treatment process was evaluated in terms of organic matter, ammonia, and manganese removal, so was its effect on the following conventional treatment process. It was found that biodegradation and filter rejection should be accounted for the pollutant removal in the hybrid processes.

2. Materials and methods

2.1. Reactor description

The schematic diagram of the hybrid process was depicted in Fig. 1, in which the pre-oxidation column and the BAF cylinder with packed media were designed in sequences. The reactors were placed in a drinking water plant located in South China.

Pre-oxidation column was made of stainless steel. Inner diameter and height of the column were 100 and 3,000 mm, respectively. Dosing solution of PPC was prepared by dissolving the solid state of PPC with tap water. The dosage of PPC was optimized as 0.6 mg/L, which was determined according to our previous tests (data not shown). Hydraulic retention time of the pre-oxidation reactor was about 25 min, and then the effluent of the pre-oxidation reactor directly flowed into the BAF reactor.

The BAF cylinder was made of plexiglass. The inner diameter and height were 100 and 3,000 mm, respectively. Up-flow mode was used throughout the whole test period, and at the bottom of this reactor, 200 mm of water distribution space in vertical

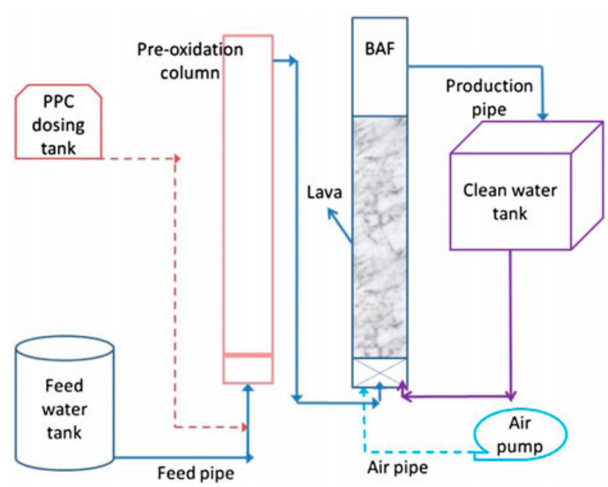

Fig. 1. Set-up of the hybrid pre-treatment strategy.

direction was designed with the perforated aeration pipe and the perforated water distribution plate. The backwashing air and water inlets were also devised. The effective height of the packed lava rock was 2,000 mm. The diameter, porosity, and density of the lava rock were 1–3 mm, 58.33% and 0.922 g/cm^3, respectively.

2.2. Experiment procedure

In fact, two identical BAF cylinders were adopted in this test with the absolutely same influent water quality. Initially, these two BAFs were operated using naturally start-up mode, with average temperature at 23℃ (20–25). Based on our previous research (data not shown), the operation conditions were fixed at 30 min of empty bed contact time, 1:1 air-to-water ratio (volume ratio, adjusted by flowmeter), and 28 L/h volumetric loading. The backwashing frequency was about twice per month.

The reactors were continuously operated for 90 d. During the first 60 d running period (about 10 d startup period and 50 d steady-state operation), the two BAFs presented almost identical results for contaminant removal. On the 61st day, the hybrid process was developed by introducing the pre-oxidation column into one of the two BAF devices as depicted in Fig. 1. The other one BAF was operated as the control experiment.

2.3. Jar test of coagulation

To examine the effect of hybrid pre-treatment process on following conventional treatment process, coagulation results of raw water and PPC-BAF effluent were compared. In jar tests, the coagulation–sedimentation test was conducted in a series of 1 L glass beakers, and the adopted coagulant was polymeric aluminum ferric chloride. The optimized dosage of coagulant for organic matter removal was 4.0 mg/L (as the mass sum of Al_2O_3 and Fe_2O_3), and the coagulation–sedimentation test was evaluated as the following procedure: adding coagulant, rapid mixing of 300 rpm for 1 min, slow mixing of 60 rpm for 10 min, and then 30 min for static sedimentation.

2.4. Raw water quality

Polluted river water, which was the drinking water source for a city in South China, was pumped as the feed in this study. The source water has been heavily polluted due to the wastewater drainage. The main qualitative properties are shown in Table 1.

Table 1
Properties of raw water

Parameters	
Temperature (℃)	20–25
pH	7.2–7.6
Turbidity (NTU)	23.5–57.2
Ammonium (mg/L)	1.00–1.60
Nitrite (mg/L)	0.16–0.33
Nitrate (mg/L)	1.69–2.94
DOC (mg/L)	5.38–7.2
UV_{254} (cm^{-1})	0.118–0.152
Manganese (mg/L)	0.16–0.73

2.5. Analytical methods

During the testing period, feed water, effluent of each unit of the hybrid process, and BAF alone were taken for the water quality analysis every day. All of the water quality analysis was conducted according to the standard methods. Ammonia, nitrite, and nitrate concentration were determined by the colorimetric methods using a spectrometer (UV721, HACH, USA); UV_{254} (pre-filtered through 0.45 μm membrane) was also determined by spectrometer. Total manganese concentration was determined with formaldoxime method using the spectrometer too. Dissolved organic matter (DOC) (pre-filtered through 0.45 μm membrane) was measured by a TOC analyzer (TOC-VCPN, Shimadzu, Japan). Turbidity was determined by a Hach 2100N turbiditymeter (Hach Company, USA). According to Standard Methods 5710B, a seven-day chlorine reaction test was employed for Disinfection By-Products Formation Potential (DBPFP) determination.

Ultrafiltration methods were adopted to determine the molecular weights distribution of organic matter, and the YM series UF membranes (100, 30, 10, 3, 1 kDa) from Millipore was adopted.

For SEM analysis, the samples were prepared following the procedures described by Mahendran et al. [17] with some modification.

2.6. Bacterial community structure analysis

Polymerase chain reaction-denaturing gradient gel electrophoresis (PCR-DGGE) had been established to analyze bacterial community structures without the inherent biases of cultivation [18]. It was adopted to monitor the microbial communities of the biomass samples from the BAF reactor in the hybrid process. The gene fragments of the microbial organisms were extracted from the BAF microbial samples and the V3 region of 16S rRNA was amplified by polymerase chain reaction (PCR). Finally, PCR products were analyzed by denaturing gradient gel electrophoresis (DGGE). The details about PCR-DGGE were presented elsewhere [19].

Shannon–Wiener Index was applied to analyze the microbial diversity of the biomass samples. Formula of Shannon–Wiener Index can be expressed as the equation below:

$$H' = -\sum_{i=1}^{S} \frac{n_i}{N} \ln \frac{n_i}{N} \quad (1)$$

where H' is the Shannon Index; S is the band number of biomass sample; n_i is the individual number of species i; N is the total individual number in a population. Here, individual number is expressed by peak area.

3. Results and discussions

3.1. Removal of pollutants

3.1.1. Removal of organic matter

For drinking water production, organic matter in raw water is always a hot research point, not only for its difficulty to be eliminated, but also due to its potential formation of disinfection byproducts [20]. The performance of BAF alone and hybrid process for the removal of DOC was examined and showed in Fig. 2(a). It could be seen that the average DOC concentration in the feed water was 7.07 mg/L, and the hybrid process could achieve 2.47 mg/L removal of DOC, with the average removal rate at 34.93%. The control test showed that BAF alone could only present 13.80% removal of DOC. Note that the feed solutions were always identical for the two processes, it could be concluded that introducing PPC oxidation into the BAF process could obviously enhance the organic matter removal.

Lower PPC dosage may only change the structure of the organic matter, rather than mineralizing the organic matter. In order to find out the mechanism for the higher DOC removal efficiency in hybrid-process than that in BAF alone, specific ultra-violet absorbance (SUVA) was determined, because it could reflect certain characteristics of the organic matter and biodegradability of the water samples [21]. SUVA, an indicator of relative unsaturated carbon content, was calculated by dividing UV_{254} by DOC. The SUVA value decreased by 0.22 L/(m mg) on average because of the PPC pre-oxidation, which indicated that PPC pre-oxidation apparently increased the biodegradability of the processing water.

Besides, the molecular weight distribution in DOC showed that PPC oxidation really increased the ratio of low molecular weight organic matter (see Fig. 2(b)), especially the part below 1 kDa, which was usually considered to be the biodegradable part of organic matter. It was shown that PPC increased <1 kDa part of organic matter from 4.30 to 5.25 mg/L, and BAF degraded <1 kDa part of organic matter to 2.79 mg/L, which was demonstrated to be lower than that after BAF filtration alone (data not shown). The fact was that though introduction of PPC with low dosage did not enhance the organic matter removal, it really changed the organic matter to be more biodegradable. The removal of organic matter would be a great reduction in organic matter loading to the following conventional drinking water process. On the other hand, the organic matter removed by PPC-BAF was relatively hydrophilic (data not shown) and with low molecular weight, which was difficult to be removed by conventional drinking water process and was a possible carbon nutrient for the bacteria in the distribution network. In this aspect, introduction of this combined pretreatment process could be an effective strategy to enhance the biosafety of drinking water.

Moreover, trihalomethane formation potential (THMFP), dichloroacetic acid formation potential (DCAAFP) and trichloroacetic acid formation potential (TCAAFP) were determined, which were reported to be the main disinfection byproducts in drinking water. As illustrated in Fig. 2(c), THMFP was observed to decrease from 267 to 166 μg/L after PPC pre-oxidation and BAF treatment, with an average removal rate of 37.83%. The average removal rates of DCAAFP and TCAAFP were 11.78 and 14.31%, respectively. Organic matter in the raw water has been demonstrated as the precursors of disinfection byproducts when chlorine was used as disinfectants [22]. As mentioned above, the hybrid process would be a good method to remove THMFP.

3.1.2. Variations of nitrogen concentration

The presence of ammonia in raw water could not only consume more disinfectant, but also cause the formation potential of nitrite in the distribution pipelines [23], which was hazardous to human health. Moreover, nitrification in the distribution pipes would reduce pH value of the drinking water, resulting in the possible release of heavy metals from pipes [24]. Thus, the removal of ammonia is significant in the whole water treatment process. The variations of ammonia, nitrite, and nitrate in the test period were determined and displayed in Fig. 3.

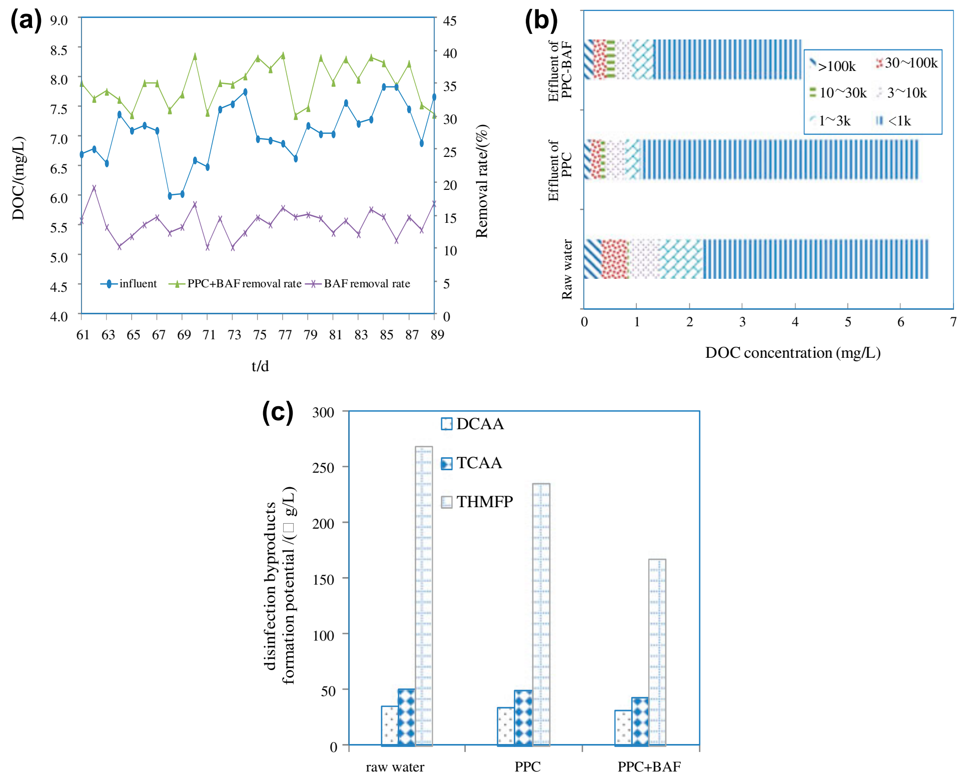

Fig. 2. Profiles about organic matter: (a) removal of DOC by the hybrid pre-treatment process, (b) molecular weights distribution and (c) profiles about disinfection by-products formation potential in the hybrid pre-treatment process.

As shown in Fig. 3(a), the average ammonium concentration in the influent was 1.64 mg/L. As for the hybrid process, PPC oxidation hardly reduced ammonia, so the removal of ammonia should be mainly attributed to the following lava-based BAF. The ammonia concentration of effluent from the hybrid process and BAF alone was both approximately 0.23 mg/L, respectively. It indicated that low dosage of PPC ahead of BAF did not affect the performance of microbes in the BAF reactor. Additionally, it was worth pointing out that the limited value of ammonium concentration in Chinese drinking water sanitary standard is 0.5 mg/L, which confirmed that BAF pre-treatment is of great efficiency.

Complete nitrification should include the effect of ammonia oxidizing bacteria and nitrite oxidizing bacteria. The former was responsible for oxidizing ammonia to nitrite, while the latter was for oxidizing nitrite to nitrate. Dissolved oxygen (DO) and pH in effluent of the hybrid process were approximately 7.2 mg/L and 7.4, respectively, which indicated that DO and pH would benefit the growth of AOB and NOB.

As illustrated in Fig. 3(b), the average nitrite concentration of influent and effluent in the hybrid process were 0.17 and 0.008 mg/L, respectively. This indicated that there was no accumulation of nitrite during the whole experiment period. It could be seen from Fig. 3(c) that the average nitrate concentrations of influent and effluent in the hybrid process were 2.50 and 3.92 mg/L, respectively. An increase in the nitrate concentration was consistent with the reduction of ammonia and nitrite, which showed that reduced ammonia and nitrite were completely oxidized to nitrate by lava-based BAF, and its performance was hardly affected by the lower PPC pre-oxidation.

3.1.3. Variation of manganese

As illustrated in Fig. 4, introduction of PPC pre-oxidation actually increased the manganese concentration in the water. Throughout the whole monitoring period, average manganese concentrations before and after PPC oxidation were 0.39 and 0.47 mg/L, respectively. Excess 0.08 mg/L of manganese was brought into the process by PPC oxidation. This was not good news for the manganese polluted raw water. However, the average manganese concentration in the effluent of BAF was only 0.05 mg/L, and the value was always below 0.1 mg/L, which meets Chinese drinking water sanitary standard. The manganese

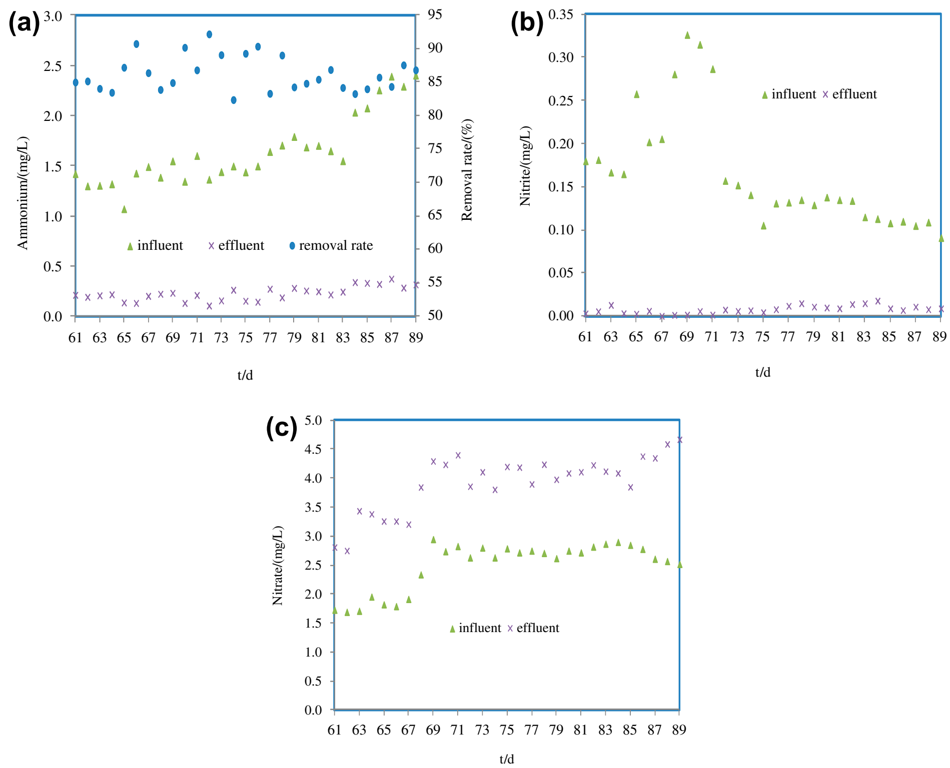

Fig. 3. Variation of nitrogen in the influent and effluent.

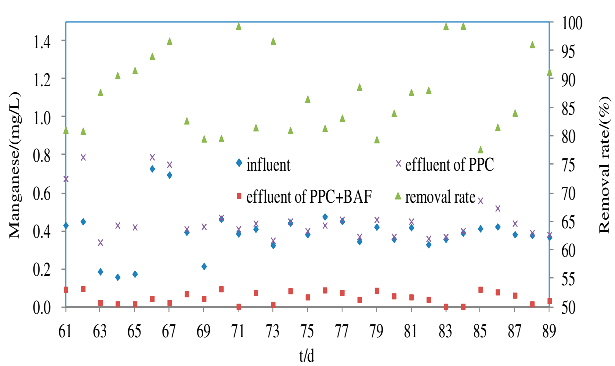

Fig. 4. Variations of manganese during the experiment period.

oxidizers or the filtering function may be accounted for the removal of manganese in BAF [11], which could obviously compensate the weakness of the PPC pre-oxidation.

When manganese oxidizing bacteria (MOB) has been immobilized and mediated the oxidation reactions of manganese, SEM could be used to observe and characterize the morphology of the exopolymers from the bacteria. Thus, at the end of this test, filters were taken out for observation by SEM, and EDS were executed. It was shown that bioflocs on the surface of the lava (see Fig. 5) had relatively more amounts of manganese. All of the elements, i.e. carbon, oxygen, phosphorous, sodium, etc. were inevitable for the microbes. In our test, it was found that the content of manganese was always around 18.06%, which may demonstrate that bioflocs were the aggregates of manganese oxidizers. Another possible explanation about this phenomenon was that manganese oxides may be precipitated on the filter surface, resulting in the formation of a coating on the surface [25].

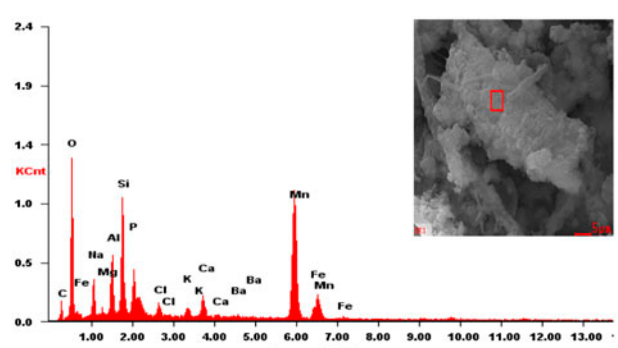

Fig. 5. SEM-EDS of the flocs on the filter surface.

3.2. SEM observation and PCR-DGGE analysis

Actually, the core of the hybrid process was the biodegradation effect of BAF. Dense biofilm has been observed on the filter surface with staggered concave–convex structure (Fig. 6). The filamentous bacteria and spherical bacteria were abounding in the salient and depression area, respectively. To demonstrate the presence of bacteria, PCR-DGGE analysis was applied to determine the bacterial communities' structure.

DGGE profile was shown in Fig. 6(b). The clusters depicted in Fig. 6(b), relating to the bands, are displayed in Table 2. DGGE fingerprints were analyzed by Smart Viewer gel imaging system, and Shannon Indexes for the biomass sample are calculated. Shannon Index is 2.43, which is similar to that in a biological process for treating wastewater reported by Wu et al. [26] and higher than other studies [27]. This demonstrated that microbial species in BAF reactor were much diversified.

For DGGE analysis, not all the bands were selected. It proved the presence of MOB, containing band *c* and *d*. The total MOB took up about 10% of the total bacteria in the detected community. *Bacillus* (see band *c* in Fig. 6(b) and Table 2) and *Pseudomonas fluorescens* (see band d in Fig. 6(b) and Table 2) were recognized as the bacteria with efficient manganese oxidizing ability [28]. Moreover, *Bacillus* and *Pseudomonas fluorescens*, belonging to Oligotrophic Bacteria [28], could adsorb organic matter and live in lower substrate concentration. Ammonia oxidizing bacteria were not detected; this may be due to the inappropriate selection of band. However, considerable mass of *Candidatus nitrospira defluvii* (see band e in Fig. 6(b) and Table 2) were found, which was reported as the dominant species as nitrite oxidizing bacteria [29].

Therefore, diversified microbial communities in the BAF reactors should be accounted for the pollutant removal stated above.

3.3. Integration of the hybrid pre-treatment process with coagulation

In order to investigate the feasibility of placing this hybrid pre-treatment process ahead of conventional treatment process, a series of jar test based on Section 2.3 were performed. In the jar tests, raw water and effluent of the hybrid process were used for coagulation experiments. The results of jar tests are detailed in Table 3.

The pre-treatment process could enhance the reduction of organic matter, increasing the removal rate of DOC and UV_{254} from 24.65 to 55.14% and from 18.70 to 30.53%, respectively. Interestingly, coagulation–sedimentation (CS) alone could hardly remove ammonia and manganese, and PPC-BAF-coagulation-sedimentation (PBCS) gave rise to higher removal of

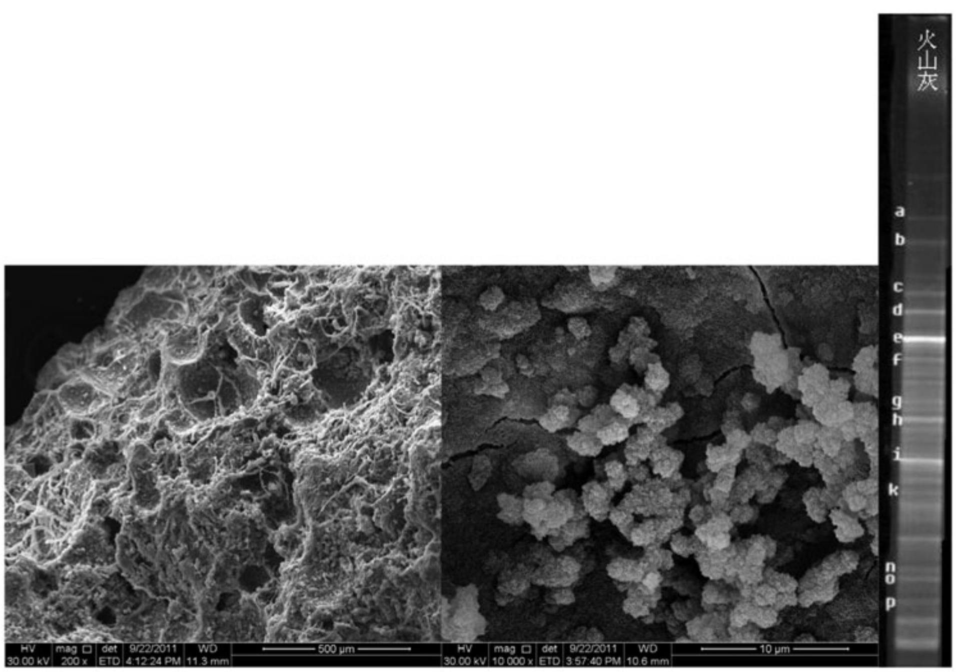

Fig. 6. (a) Morphology of filter surface captured by SEM (200× and 10,000×, respectively) and (b) denaturing gradient gel electrophoresis profiles of bacteria gene in the BAF reactor.

Table 2
Bacteria determined to be responsible for impurities removal

Band	V3 region	Database serial number	Strain	Gray percentage (%)
c	116 bp	AY277557	*Bacillus weihenstephanensis*	1.62
d	193 bp	AJ308308	*Pseudomonas fluorescens*	4.32
e	183 bp	DQ059545	*Candidatus nitrospira defluvii*	26.11
g	160 bp	AY986507	*Bacillus cereus*	2.71

Table 3
Results about the effects of hybrid pre-treatment methods on the following coagulation

Process	Removal rate (%)				Effluent
	DOC	UV_{254}	Ammonium	Manganese	Turbidity (NTU)
CS[a]	24.65	18.70	8.84	6.29	1.56
PBCS[b]	55.14	30.53	90.13	87.24	0.72

[a]Coagulation and sedimentation.
[b]PPC/BAF/coagulation/sedimentation.

ammonia and manganese at 90.13 and 87.24%, respectively. Moreover, effluent of PBCS presented a lower turbidity than that of CS [30,31]. Except for particles and organic matter, it was reported that conventional treatment with biological pre-filtration could effectively improve the removal of odorants, such as geosmin and MIB [31].

Actually, PPC could react with many inorganic and organic matters in water samples, and the main component in PPC was reported as potassium permanganate. During the reaction process of PPC in water hydrous manganese dioxide could be generated. The forming hydrous manganese dioxide could provide reactive surface sites and showed excellent adsorption characteristics [32]. Hydrous manganese dioxide could enhance the heterogeneous coagulation process and facilitate the formation of larger floc by acting as nucleating sites [33].

This hybrid pretreatment process did not negatively influence the following coagulation-sedimentation efficiency; it could remove biodegradable organic matter and ammonia, which would be great problems to the conventional treatment process. Therefore, its success would greatly reduce the pollutant loading and the post-chlorine dosage, thus improve the drinking water safety.

3.4. Implications for practice

Pollutants including ammonia, organic matter, and manganese had been considered as the new pollution problem. In some developing countries, e.g. China, with the increasing economic development, more and more water resources are being or have been polluted, which would compromise the efficiency of conventional drinking water treatment processes. Though integrating biological pretreatment into an old conventional treatment line would bring considerable investment cost and operating cost, the development of pre-treatment processes will be of great interest to the drinking water engineers.

As stated above, hybrid pre-treatment process presented a removal rate of organic matter at 34.92%, the significance was that if not, the removal part would heavily influence the following treatment process and/or the water quality in the distribution network. Moreover, it is important to control ammonia concentration by the biological method rather than chemical method, which would usually produce much byproduct. Combining this effective method with the conventional drinking water would not only greatly improve the stability of drinking water works, but also could increase the grade of safe drinking water quality. In some specific cases, the advantages of this hybrid pretreatment (increased particle and DOC removal, ammonia and manganese reduction) should be weighed against the costs when compared to other upgrading process such as membrane filtration [34].

4. Conclusions

By treating polluted source water, continuous running of hybrid pre-treatment process was evaluated. As expected, PPC pre-oxidation could increase the biodegradability of raw water, resulting higher removal of DOC by the following BAF. Besides, introduction of PPC presented no inhibition on the nitrifiers in BAF during the whole experimental period. PPC could increase the manganese concentration, but the average concentration of manganese in effluent of the following BAF was 0.05 mg/L; PBCS could apparently achieve higher removal of contaminants than CS. Furthermore, it was demonstrated that the removal of pollutants should be attributed to the bioflocs on the lava-filter surface. The pretreatment process would be of great significance due to its removal of biodegradable organic matter and ammonia for biosafety improvement and be of guiding significance to the promotion and operation of drinking water works. However, if the BAF did not work, biodegradable DOC could enter the distribution systems, which may induce the growth potential of bacteria in pipes [35]. Thus, optimization of organic matter removal and risk evaluation should be further studied and improved.

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